

## CLAIMS

1. A method of manufacturing a solid oxide fuel cell module comprising a plurality of cells each made up of a fuel electrode, an electrolyte, and an air electrode sequentially formed on a surface of a substrate with an internal fuel flow part provided therein, at least a face of the substrate, in contact with the cells, and interconnectors, being an insulator, and the cells adjacent to each other, being electrically connected in series through the intermediary of the respective interconnectors,

    said method of manufacturing the solid oxide fuel cell module comprising the steps of:

        co-sintering the respective fuel electrodes, and the respective electrolytes;

        subsequently forming a dense interconnector out of a dense interconnector material, or an interconnector material turning dense by sintering in at least parts of the solid oxide fuel cell module, in contact with the respective fuel electrodes, and the respective electrolyte; and

        forming an air electrode on the respective electrolytes before electrically connecting the air electrode with the respective dense interconnectors.

2. A method of manufacturing a solid oxide fuel cell module comprising a plurality of cells each made up of a fuel electrode, an electrolyte, and an air electrode sequentially formed on a surface of a substrate with an internal fuel flow part provided therein, at least a face of the substrate, in contact with the cells, and interconnectors, being an insulator, and the cells adjacent to each other, being electrically connected in series through the intermediary of the respective interconnectors,

    said method of manufacturing the solid oxide fuel cell module comprising the steps of:

        co-sintering the substrate, the respective fuel electrodes, and the respective electrolytes;

        subsequently forming a dense interconnector out of a dense interconnector material, or an interconnector material turning dense by sintering in at least parts of the solid oxide fuel cell module, in contact with

the respective fuel electrodes, and the respective electrolytes; and

forming an air electrode on the respective electrolytes before electrically connecting the air electrode with the respective dense interconnectors.

3. A method of manufacturing a solid oxide fuel cell module comprising a plurality of cells each made up of a fuel electrode, an electrolyte, and an air electrode sequentially formed on a surface of a substrate with an internal fuel flow part provided therein, at least a face of the substrate, in contact with the cells, and interconnectors, being an insulator, and the cells adjacent to each other, being electrically connected in series through the intermediary of the respective interconnectors,

said method of manufacturing the solid oxide fuel cell module comprising the steps of:

co-sintering the respective fuel electrodes, the respective electrolytes, and a dense interconnector material, or an interconnector material turning dense by co-sintering, in at least parts of the solid oxide fuel cell module, in contact with the respective fuel electrodes, and the respective electrolytes; and

forming an air electrode on the respective electrolytes before electrically connecting the air electrode with the respective dense interconnectors.

4. A method of manufacturing a solid oxide fuel cell module comprising a plurality of cells each made up of a fuel electrode, an electrolyte, and an air electrode sequentially formed on a surface of a substrate with an internal fuel flow part provided therein, at least a face of the substrate, in contact with the cells, and interconnectors, being an insulator, and the cells adjacent to each other, being electrically connected in series through the intermediary of the respective interconnectors,

said method of manufacturing the solid oxide fuel cell module comprising the steps of:

co-sintering the substrate, the respective fuel electrodes, the respective electrolytes, and a dense interconnector material, or an interconnector material turning dense by co-sintering, in at least parts of the

solid oxide fuel cell module, in contact with the respective fuel electrodes, and the respective electrolytes; and

forming an air electrode on the respective electrolytes before electrically connecting the air electrode with the respective dense interconnectors.

5. A method of manufacturing a solid oxide fuel cell module comprising a plurality of cells each made up of a fuel electrode, an electrolyte, and an air electrode sequentially formed on a surface of a substrate with an internal fuel flow part provided therein, at least a face of the substrate, in contact with the cells, and interconnectors, being an insulator, and the cells adjacent to each other, being electrically connected in series through the intermediary of the respective interconnectors,

said method of manufacturing the solid oxide fuel cell module comprising the steps of:

disposing a dense interconnector material, or an interconnector material turning dense by co-sintering, in portions of the respective fuel electrodes;

subsequently covering the respective fuel electrodes, and the dense interconnector material, or the interconnector material turning dense by co-sintering before co-sintering the respective fuel electrodes, the dense interconnector material, or the interconnector material turning dense by co-sintering, and the respective electrolytes, thereby forming dense interconnectors;

forming an air electrode on the respective electrolytes; and

subsequently electrically connecting the air electrode with the respective dense interconnectors.

6. A method of manufacturing a solid oxide fuel cell module comprising a plurality of cells each made up of a fuel electrode, an electrolyte, and an air electrode sequentially formed on a surface of a substrate with an internal fuel flow part provided therein, at least a face of the substrate, in contact with the cells, and interconnectors, being an insulator, and the cells adjacent to each other, being electrically connected in series through the intermediary of the respective interconnectors,

said method of manufacturing the solid oxide fuel cell module comprising the steps of:

disposing a dense interconnector material, or an interconnector material turning dense by co-sintering, in portions of the respective fuel electrodes;

subsequently covering the respective fuel electrodes, and the dense interconnector material, or the interconnector material turning dense by co-sintering before co-sintering the substrate, the respective fuel electrodes, the dense interconnector material, or the interconnector material turning dense by co-sintering, and the respective electrolytes, thereby forming dense interconnectors;

forming an air electrode on the respective electrolytes; and

subsequently electrically connecting the air electrode with the respective dense interconnectors.

7. A method of manufacturing a solid oxide fuel cell module according to any of claims 1 to 6, wherein a mixture of MgO, and MgAl<sub>2</sub>O<sub>4</sub> is used as a constituent material of the substrate with the internal fuel flow part provided therein, at least a face of the substrate, in contact with the cells, and the interconnectors, being the insulator.

8. A method of manufacturing a solid oxide fuel cell module according to claim 7, wherein the mixture of MgO, and MgAl<sub>2</sub>O<sub>4</sub> is a mixture of MgO, and MgAl<sub>2</sub>O<sub>4</sub>, containing 20 to 70 vol. % of MgO.

9. A method of manufacturing a solid oxide fuel cell module according to any of claims 1 to 6, wherein an yttria stabilized zirconia expressed by chemical formula (Y<sub>2</sub>O<sub>3</sub>)<sub>x</sub> (ZrO<sub>2</sub>)<sub>1-x</sub> (in the chemical formula, x = 0.03 to 0.12) is used as a constituent material of the substrate with the internal fuel flow part provided therein, at least a face of the substrate, in contact with the cells, and the interconnectors, being the insulator.

10. A method of manufacturing a solid oxide fuel cell module according to any of claims 1 to 6, wherein a mixture of a mixture composed of MgO, and MgAl<sub>2</sub>O<sub>4</sub>, and an yttria stabilized zirconia expressed by chemical formula (Y<sub>2</sub>O<sub>3</sub>)<sub>x</sub> (ZrO<sub>2</sub>)<sub>1-x</sub> (in the chemical formula, x = 0.03 to 0.12) is used as a constituent material of the substrate with the internal fuel flow part

provided therein, at least a face of the substrate, in contact with the cells, and the interconnectors, being the insulator.

11. A method of manufacturing a solid oxide fuel cell module according to claim 10, wherein the mixture of MgO, and MgAl<sub>2</sub>O<sub>4</sub> is a mixture of MgO, and MgAl<sub>2</sub>O<sub>4</sub>, containing 20 to 70 vol. % of MgO.

12. A method of manufacturing a solid oxide fuel cell module according to any of claims 1 to 11, wherein a constituent material of the substrate with the internal fuel flow part provided therein, at least a face of the substrate, in contact with the cells, and the interconnectors, being the insulator, is material composed of Ni diffused in a range not more than 35 vol. %.

13. A method of manufacturing a solid oxide fuel cell module according to any of claims 1 to 12, wherein material composed mainly of Ni, is used as a constituent material of the fuel electrode.

14. A method of manufacturing a solid oxide fuel cell module according to any of claims 1 to 12, wherein a mixture of Ni and an yttria stabilized zirconia expressed by chemical formula (Y<sub>2</sub>O<sub>3</sub>)<sub>x</sub>(ZrO<sub>2</sub>)<sub>1-x</sub> (in the chemical formula, x = 0.03 to 0.12), with not less than 40 vol. % of Ni diffused in the mixture, is used as a constituent material of the fuel electrode.

15. A method of manufacturing a solid oxide fuel cell module according to any of claims 1 to 14, wherein, an yttria stabilized zirconia expressed by chemical formula (Y<sub>2</sub>O<sub>3</sub>)<sub>x</sub>(ZrO<sub>2</sub>)<sub>1-x</sub> (in the chemical formula, x = 0.05 to 0.15) is used as a constituent material of the electrolyte.

16. A method of manufacturing a solid oxide fuel cell module according to any of claims 1 to 14, wherein a scandia stabilized zirconia expressed by chemical formula (Sc<sub>2</sub>O<sub>3</sub>)<sub>x</sub>(ZrO<sub>2</sub>)<sub>1-x</sub> (in the chemical formula, x = 0.05 to 0.15) is used as a constituent material of the electrolyte.

17. A method of manufacturing a solid oxide fuel cell module according to any of claims 1 to 14, wherein an yttria doped ceria expressed by chemical formula (Y<sub>2</sub>O<sub>3</sub>)<sub>x</sub>(CeO<sub>2</sub>)<sub>1-x</sub> (in the chemical formula, x = 0.02 to 0.4) is used as a constituent material of the electrolyte.

18. A method of manufacturing a solid oxide fuel cell module according to any of claims 1 to 14, wherein a gadolinia doped ceria expressed

by chemical formula  $(\text{Gd}_2\text{O}_3)_x(\text{CeO}_2)_{1-x}$  (in the chemical formula,  $x = 0.02$  to  $0.4$ ) is used as a constituent material of the electrolyte.

19. A method of manufacturing a solid oxide fuel cell module according to any of claims 1 to 18, wherein material composed of a mixture of a glass and an electroconductive material is used as a constituent material of the interconnector.

20. A method of manufacturing a solid oxide fuel cell module according to claim 19, wherein the glass in the mixture of the glass and the electroconductive material is a glass with thermal expansion coefficient falling in a range of  $8.0$  to  $14.0 \times 10^{-6} \text{ K}^{-1}$ .

21. A method of manufacturing a solid oxide fuel cell module according to any of claims 19 to 20, wherein the glass in the mixture of the glass and the electroconductive material is a glass with a softening point falling in a range of  $600$  to  $1000^\circ\text{C}$ .

22. A method of manufacturing a solid oxide fuel cell module according to any of claims 19 to 21, wherein the electroconductive material in the mixture of the glass and the electroconductive material is a metal.

23. A method of manufacturing a solid oxide fuel cell module according to claim 22, wherein the metal is at least one kind of metal selected from the group consisting of Pt, Ag, Au, Ni, Co, W, and Pd.

24. A method of manufacturing a solid oxide fuel cell module according to claim 22, wherein the metal is an alloy containing Ag.

25. A method of manufacturing a solid oxide fuel cell module according to any of claims 19 to 21, wherein the electroconductive material in the mixture of the glass and the electroconductive material is an electroconductive oxide.

26. A method of manufacturing a solid oxide fuel cell module according to claim 25, wherein the electroconductive oxide is a perovskite type ceramics composed of not less than two elements selected from the group consisting of La, Cr, Y, Ce, Ca, Sr, Mg, Ba, Ni, Fe, Co, Mn, Ti, Nd, Pb, Bi, and Cu.

27. A method of manufacturing a solid oxide fuel cell module according to claim 25, wherein the electroconductive oxide is an oxide

expressed by chemical formula (Ln, M) CrO<sub>3</sub> (in the chemical formula, Ln refers to lanthanoids, and M refers to Ba, Ca, Mg, or Sr).

28. A method of manufacturing a solid oxide fuel cell module according to claim 25, wherein the electroconductive oxide is an oxide expressed by chemical formula M (Ti<sub>1-x</sub> Nb<sub>x</sub>) O<sub>3</sub> (in the chemical formula, M refers to at least one element selected from the group consisting of Ba, Ca, Li, Pb, Bi, Cu, Sr, La, Mg, and Ce, x = 0 to 0.4).

29. A method of manufacturing a solid oxide fuel cell module according to any of claims 19 to 28, wherein electroconductive material content of the mixture of the glass and the electroconductive material is not less than 30 vol. % of the mixture.

30. A method of manufacturing a solid oxide fuel cell module according to any of claims 19 to 29, wherein the mixture of the glass and the electroconductive material is subjected to heat treatment at not higher than the melting point of the electroconductive material after the mixture is applied between the fuel electrode of one of the adjacent cells, and the air electrode of the other cell.

31. A method of manufacturing a solid oxide fuel cell module according to any of claims 1 to 29, wherein only portions of the interconnector connecting the fuel electrode of one of the adjacent cells with the air electrode of the other cell, in contact with the fuel electrode, and the electrolyte, respectively, are formed of material composed mainly of Ag.

32. A method of manufacturing a solid oxide fuel cell module according to any of claims 1 to 29, wherein only portions of the interconnector connecting the fuel electrode of one of the adjacent cells with the air electrode of the other cell, in contact with the fuel electrode, and the electrolyte, respectively, are formed of material composed of one kind or not less than two kinds of material selected from the group consisting of Ag, Ag solder, and a mixture of Ag and the glass.

33. A method of manufacturing a solid oxide fuel cell module according to any of claims 1 to 29, wherein only portions of the interconnector connecting the fuel electrode of one of the adjacent cells with the air electrode of the other cell, in contact with the fuel electrode, and the

electrolyte, respectively, are formed of an electroconductive oxide.

34. A method of manufacturing a solid oxide fuel cell module according to any of claims 1 to 29, wherein an oxide material containing Ti is used as a constituent material of the interconnector connecting the fuel electrode of one of the adjacent cells with the air electrode of the other cell,

35. A method of manufacturing a solid oxide fuel cell module according to claim 34, wherein the oxide material containing Ti is material expressed by chemical formula  $M(Ti_{1-x}Nb_x)O_3$  (in the chemical formula, M refers to at least one element selected from the group consisting of Ba, Ca, Pb, Bi, Cu, Sr, La, Li, and Ce,  $x = 0$  to  $0.4$ )